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DATA ASSESSMENT REPORT

Technical Directive: 05
Technical Directive Title: Reilly Tar Technical Support

Technical Directive Report

by

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for

ICAIR SYSTEMS DIVISION LIFE SYSTEMS, INC. 24755 Highpoint Road Cleveland, OH 44122 This report is divided into three sections. Coal tar distillation, the constituents of coal tar and the formation and degradation of PAHs are discussed in the first section. In the second section W13, W23, SLP4 and SLP15 are assessed as to their levels of PAHs present in the water. In the third section recommendations for further work are presented.

SECTION I

1. Source

Coal Tar Production

Coal can be described as a compact stratified mass of vegetation, interspersed with smaller amounts of inorganic matter, which has been modified chemically and physically by agents over time. These agents include the action of bacteria and fungi, oxidation, reduction, hydrolysis and condensation, and the effects of heat and pressure in the presence of water. The chemical properties of coal depend upon the amounts and ratios of different constituents present in the vegetation, as well as the nature and quantity of inorganic material and the changes which these constituents have undergone (Francis, 1961).

. Coal, therefore, has a rather complicated chemical structure based on carbon and hydrogen with varying amounts of oxygen, nitrogen and sulfur. <u>Bituminous coal</u>, from which coal tar and coal tar pitch are derived, contains a number of PAHs, including carcinogenic benzo(a)pyrene (BaP) and benz(a)anthracene (Tye et al., 1966), and a variety of toxic trace elements such as antimony, arsenic, beryllium, cadmium, lead, nickel, chromium, cobalt, titanium, and vanadium (Zubovic, 1975).

When coal is pyrolyzed, a variety of changes occur: above 100°C free water evaporates; above 200°C combined water and carbon dioxide are evolved; above 350°C bituminous coals soften and melt, decomposition begins, and tar and gas are evolved; at 400 to 500°C most of the tar is evolved; at 450 to 550°C decomposition continues and the residue turns solid; above 550°C the solid becomes coke and only gas is evolved; around 900°C no more gas is evolved and only coke remains; above 900°C small physical changes occur.

When coal undergoes carbonization, it passes through two steps of decomposition: onset of plasticity at 350 to 500°C and advanced decomposition at 650 to 750°C. Volatile products released at each stage undergo a series of secondary reactions as they pass through the coke before emerging from the retort. The volatiles are separated by fractional condensation or absorption into tar, ammoniacal liquor, benzole, and illuminating or heating gas (McNeil, 1966a).

The major reactions in the conversion of primary carbonization products into tars (McNeil, 1966a) are:

- cracking of higher molecular weight paraffins to gaseous paraffins and olefins;
- dehydrogenation of alkylcyclic derivatives to aromatic hydrocarbons and phenols;
- 3) dealkylation of aromatic, pyridine and phenol derivatives;

- 4) dehydroxylation of phenols;
- 5) synthesis of PAH by condensation of simpler structures;
- 6) disproportionation of PAH to both simpler and more complex structures.

The temperature of carbonization and contact time with the hot coke bed and heated walls of the retort will determine the composition of tars, as well as the extent of the reactions. Tars from the different types of carbonization processes vary widely as to their composition and characteristics. The term low temperature carbonization refers to pyrolysis of coal to a final temperature of 700°C. The final solid product is a weak coke with high yields of tar and oil and low yield of gas. High temperature carbonization is pyrolysis of coal between 900°C and 1200°C, with town gas as the product and coke and the byproduct at the lower temperature and metallurgical coke as the product and gas as the by-product at the higher temperature (Encyclopaedia Britannica, 1974).

Coal tar pitch is the residue from the processing of coal tar (Figures 1 and 2). Pitches or "refined tars" are obtained from the distillation of tars and represent from 30 to 60% of the tar components and commercial creosote: represents from 30-55% of the tar components. (McNeil, 1966a) (Table 1).

McNeil (1966a) has described the change in composition of tars found as the temperature increases from vacuum distillation or low temperature carbonization to high temperature carbonization of coal (Table 1):

- a) The amounts of paraffins and naphthenes decrease and disappear, the naphthenes fading out before the paraffins.
- b) The amount of phenolic material falls from about 30% to a small value.
- c) The proportion of aromatic hydrocarbons increases from alow figure to over 90%.
- d) The proportions of aromatic, phenolic and heterocyclic compounds containing alkyl side chains decrease markedly.
- e) The proportion of condensed ring compounds containing more than three fused rings increases.
- f) The yield of coal carbonized decreases from 10% to less than 5%.

Two major objectives in the primary distillation of coal tar are to obtain a pitch or refined tar residue of the desired softening points and to concentrate as far as possible in certain fractions those components which are subsequently recovered. In the case of coke oven tar distillation (Table 2) the major objective in this situation was to concentrate and blend the right and heavy oils into creosote.

2. Coal Tar Composition and Distillation

The composition of coal tar by boiling point ranges are given in Tables 3 and 4. As you will note, the size and structure of the compounds increase with the boiling range. The relative amounts of the major components of the lower boiling compounds present in the light and heavy oils are indicated in Table 5. From the primary fractions of distilling up to 160°C, crude benzole and light oil are distilled. The major components of these fractions include benzene, toluene, mixed xylenes and a residue to naphtha. From primary fractions distilling in the 159-200°C range, a series of pyridine bases as well as solvent and heavy naphthas and coumarine resins are distilled. The major components of these fractions include pyridine and the methyl pyridines, solvent naphtha and heavy naphtha and indene resins. From the primary fractions distilling in the range 200-250°C, tar acids and naphthaline oils are distilled. The tar acids include phenol, o-cresol, m-cresol and p-cresol and naphthalene is recovered from the naphthalene oil.

No tar chemicals are extracted commercially from oil in the distilling range 250-300°C. Most of these oils are used in greosote blends. From the primary fractions distilling in the range 300-350°C, anthracene, phenanthrene and carbazole are recovered. The remainder of the coal tar is a pitch which contains most of the heavier polycyclic aromatic hydrocarbons (PAHs). (Table 6). It should be noted that compounds, such as fluorene, phenanthrene and anthracene, fluorathene, pyrene, benzofluorenes, benz(a)anthracene, chrysene and triphenylene, benzo(k)fluoranthene, benzo(a)pyrene and benzo(e)pyrene, benzo(b)chrysene and o-phenylenepyrene, and benzo(ghi)perylene and anthanthrene are present in the largest amounts. PAHs can also be present in the anthracene and heavy oils due to the variability in collection of the various boiling range fractions.

The residual distillate oils after they have been processed for the extraction of naphthalene, anthracene, tar acids and tar bases, can be blended with pitch or base tar to yield various grades of creosote (Table 2). A creosote may therefore be defined as a blend of coal tar distillate oils from which more valuable components have been more or less extracted, formulated to meet particular specifications. On the other hand, creosote may be a blend of anthracene, naphthalene and heavy oils and pitch without prior extraction of the oils.

The PAHs present in commercial creosote and coal tar are compared in Table 7. The PAHs present in the largest quantities in both coal tar and creosote are similar to that shown in Table 6. i.e. anthracene, fluoranthene and phenanthrene. More importantly, it should be noted that the PAHs are present in creosote in significantly larger quantities than in coal tar. This is due to the fact that the distillation of coal tar will concentrate PAHs in the heavier oils and pitch. Therefore, these observations are consistent with the blending of heavier oils with pitch to produce creosote.

3. PAHs in the Environment

Human exposure to PAHs from a variety of sources has been estimated (Table 8a). Exposure from water is low in relation to food. Impact of total PAH and BaP to the aquatic environment has also been estimated (Table 8b). However, it should be noted that the quality of monitoring data available for developing these estimates varies considerably, and the data obtained from the drinking water were based upon six representative PAHs. three of which have carcinogenic activity, benzo(a)-pyrene, benzo(b)fluoranthene and indeno(1,2,3-c,d)pyrene. The other three are

fluoranthene, benzo(ghi)perylene and benzo(k)fluoranthene. These were chosen originally because of the case with which PAHs could be analyzed and not based on their toxicological properties. Also, BaP is not necessarily a good indicator compound. Most of what we know about ground water is based upon these analyses (Table 9-12). There is one report on PAHs in Belgium aquifers (Quaghebeur and DeWulf, 1978) using similar analyses.

There is data available on PAHs in ground water and surface water that include more carcinogenic PAHs (Tables 13 and 14) and give a better representation of the potential hazards present. The carcinogenic PAHs include benzo(a)pyrene, benzo(j)fluoranthene, benzo(b)fluoranthene, indeno(1,2,3-c,d)pyrene, benz(a)-anthracene and dibenz(a,h)anthracene. It should be noted that exposure to PAHs, from water is an involuntary exposure whereas food is more of a voluntary exposure based on social and cultural habits. Since drinking water is a municipal source, the levels of PAHs must be kept at a minimum and, therefore, 2.8 ng/l for a carcinogenic PAH, 28 ng/l for all carcinogenic PAHs and, in particular, 280 ng/l for all PAHs may in fact may be too high for human exposure.

4. PAH Decomposition and Removal

PAHs can be chemically degraded by environmental oxidizers such as ozone, chlorine, singlet oxygen, organic and hydroxyl radicals (Table 15 & 16). The particular degradations of PAHs are dependent on the chemical oxidizers used. For example, organic radicals will degrade anthracene faster than benzo(a)pyrene, but they have similar degradation properties in the presence of singlet oxygen.

In the presence of natural light smaller PAHs will decompose much faster than larger PAHs (Figure 17). After 10 hours of exposure almost 70% of compounds, such as anthracene and phenanthrene and 100% of benz(a)anthracene have been photodegraded, while compounds such as fluorene, pyrene, and benzo(a)pyrene and chrysene remain intact.

The more fused rings there are in the molecular structure of the PAHs, the more photochemically stable the compounds appear to be.

PAHs can also be degraded by microorganisms (Table 18). As you will note the degradation of benzo(a)pyrene is very slow in comparison with naphthalene. Most of what is known on the degradation of PAHs is related to the work on naphthalene. Phenols are also biodegraded rapidly while other compounds, such as aromatic amines and thiopenes are biodegraded slowly (Herbes et al.). It would appear that the smaller atomic ring molecules and cata-condensed aromatic compounds (no carbon atom belongs to more than two rings) are biodegraded more readily in the environment.

Lastly, PAHs can be removed by conventional treatments, such as flocculations, sedimentation, and filtration in addition to the methods just described (Sorrell et al., 1980, Table 19). A major factor in the reduction of PAHs are their sorption properties of PAHs and their ability to be removed by adsorption onto sediment and particulate matter. The role of chlorination is not clear since the products of chlorination may be hazardous themselves. Activated carbon can further assist in PAH removal.

SECTION II

The measured constitutents in W13 and W23 are shown in Tables 20 and 21. The tar sample in W23 contains similar compounds to that found in coal tar (lable 3,4,6) and in fairly large concentrations. In particular, the relative composition of phenanthrene, fluoranthene, pyrene and benzo(b)fluoranthene to the rest of the PAHs in the mixture are in very good agreement with that seen in coal tar (Table 6) and therefore it can be stated that tar is coal tar. The water samples from W23 contain higher amounts of naphthalene than the other PAHs due to the fact that naphthalene a two ring compound is more soluble in water than the three ring compounds. The smaller PAHs are leached from the tar more readily than compounds, such as fluorene, chrysene, benzo(a)pyrene and benzo(b)fluoranthene. Since it is known that the carcinogenic PAHs are usually 4 to 6 rings in size, these PAHs will not be found as readily in aqueous samples as the more soluble, smaller ring hon-carcinogenic compounds. It should be noted that only two carcinogenic PAHs have been analyzed for in W23 i.e. benzo(a)pyrene and benzo(b)fluoranthene.

W13 has been analyzed for both PAHs, heterocyclic analogs and aromatic amines (Table 21). It is quite clear that the water has been contaminated by PAHs from coal tar, as in the case of W23. This well has been analyzed for four carcinogenic PAHs, benzo(a)pyrene, benz(a)anthracene, dibenz(a,h)anthracene, and indeno(1,2,3-c,d) pyrene, in addition to a number of heterocyclic analogs. The carcinogenic PAHs are present in extremely high amounts. In general, compounds that are present in relative significant amounts in comparison with the other PAHs, i.e. fluoranthene and pyrene, are present in ratios similar to that found in coal tar (Table 6). It can be stated without any uncertainty that the profile of compounds represents products from coal tar.

The PAHs present in SLP4 and 15 are listed in Tables 22 and 23. In both cases it appears that more PAHs are being leached into the water system with time. It also can be stated that the carcinogenic PAHs are present in the wells in very small quantities with respect to the noncarcinogenic PAHs. The carcinogenic and/or larger PAHs may not be present in these wells in large quantities due to their lower solubilities in water. These PAHs would then tend to precipitate out of solution and be adsorbed on particulate and sediment. The smaller PAHs, on the other hand, will tend to undergo chemical or biodegradation more readily, and therefore, lower the amounts of these PAHs in well water.

It should be noted that the levels of PAHs in both wells exceed the ambient water quality standards. The types of compounds present in the wells are the same types found in coal tar.

SECTION III

The following recommendations are in order:

1) More carcinogenic PAHs should be analyzed for in the wells in addition to:

benzo(a)pyrene
benz(a)anthracene
benzo(ghi)perylene & indeno(1,2,3,-c,d)pyrene

These should be the following:

benzo(c)phenanthrene benzo(b)fluoranthene benzo(j)fluoranthene benzo(j)aceanthrylene dibenzo(a,h)pyrene dibenz(a,i)pyrene dibenz(a,e)pyrene dibenzo(c,g)carbazole dibenz(a,j)acridine dibenz(a,h)acridine

- 2) The wells should be <u>analyzed</u> for aromatic <u>amines</u> due to the lack of oxygen present, therefore, these compounds may be very stable. Compounds should be picked from these listed in Table 6. Compounds would include 1- and 2-naphthylamine, 6-amino chrysene and 2-aminoanthracene, as well as quinoline and indole type compounds.
- Some cocarcinogenic compounds are analyzed for including pyrene, fluoranthene, and benzo(ghi)perylene. Since these have biological activities enhancing the carcinogenicity of known carcinogens the following compounds as such should be included: (Slaga et al., 1980)

phenol derivatives catchol pyrogallol decane indecane tetradecane n-dodecane benzo(e)pyrene

4) Biological testing on well water samples should be undertaken. This will give some indication as to the potential biological hazard associated with the mixture. Salmonella/microsome (Ames) assay is considered the best overall assay for assessing hazards of mixtures. It is accurate to within 90%. Those which are negative in this assay should be analyzed by a second round of assays which will complement the Ames assay to pick up false negatives. These include unscheduled DNA synthesis, sister chromatid exchange and possibly a transformation assay such as hamster kidney cells, C3H/101½ cells or 3T3 cells. The hamster kidney cells assay is the easier assay to use. If too many assays are used and not for a specific test then false positives will be incorporated into the results. The results of these assays can then be used to better assess the analytical data and determine how far from the source there may still be potential hazards. (deSerres and Ashby, 1981)

- The analyses for SLP4 and 1S have been performed over times by different analytical laboratories by either GC-MS and/or HPLC. In the future it would be more appropriate if the <u>same samples</u> are analyzed by at least two different laboratories by the same techniques for quality control. It is difficult to relate the results of one laboratory with a second laboratory if the analyses are not performed on the same samples over time. It is also important if each laboratory would receive a known standard mixture of PAHs and other compounds so that the agency itself can assess the quality of work of every laboratory performing analytical services. The same procedures should also be used for biological testing.
- 6) Based on the results, there appears to be <u>anaerobic degradations of phenols</u> and <u>small PAHs</u>. There is very little information in the literature on this type of biodegradation. Most of what is known about biodegradations is based on aerobic degradations. <u>More work needs to be done in this area</u>.
- 7) A critical review on the need for control of airborne fine particulate benzo(a)pyrene has been published (Perera, 1981). The same type of review is necessary for the control of PAHs and benzo(a)pyrene in drinking water. The EPA water quality document is based on total PAHs and BaP in the water. What is more important, is whether BaP is a good indicator compound for contaminated water. BaP in fact may not be appropriate. In the past BaP has been chosen because of its ease of detectability and not based on its appropriateness as an indicator compound for contaminated water. This needs to be assessed more critically in the future.

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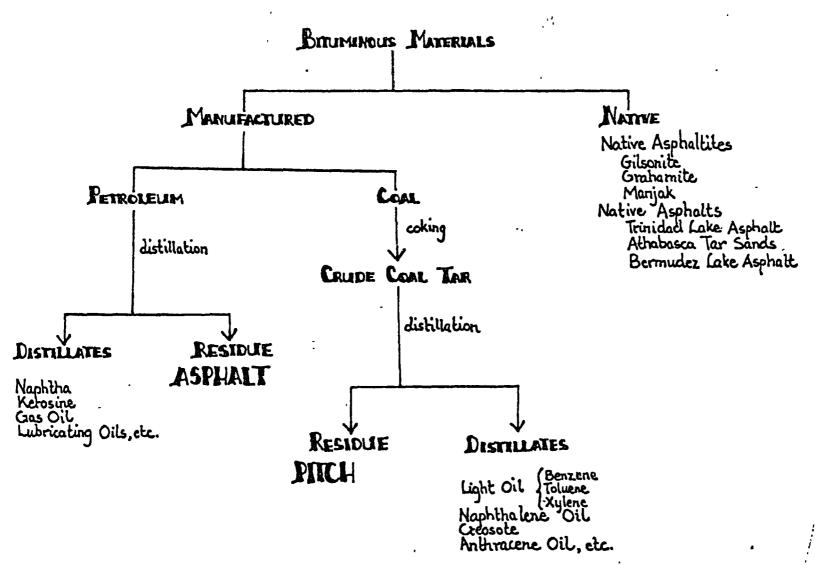
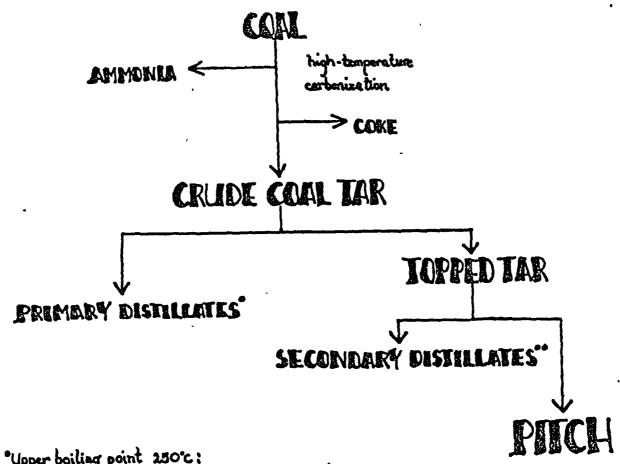


FIGURE .]. PARTIAL CLASSIFICATION OF BITUMINOUS MATERIALS



*Upper boiling point 250°C:
Includes benzine, toluene, xylene, naphtha and commarone.
*Includes onthrocene and creosote fractions.

FIGURE 2. ORIGIN OF COAL TAR PITCH

Table 1
Typical Analysis (Percent by Weight) of Tars

D.S. J.	Coke Oven Tar	Gas Works Tar	Low Temperature Tar (200°C)
Pitch	59.0		iar (200°C)
Creosote		44.0	26.0
-	31.0	42.0	rr a
Light Oils	2.5		55.0
Heavy Oils		5.4	6.7
J	5.4	6.5	9.4

Source: Encyclopaedia Britannica, 1974.

Table 2

TYPICAL FRACTIONS TAKEN IN CONTINUOUS TAR DISTILLATION TO MEDIUM-SOFT PITCH

		Coke Oven	
Fraction No.	Names	Boiling Range ^{a O} C	Wt. % of Crude Tar
1	crude benzole, light oil	99-160	0.6
2	naphtha, light oil	168-196	2.9
3	naphthalene oil	198-230	14.6
4	wash oil, benzole absorbing oil, light creosote	224-286	2.8
5	anthracene oil, heavy creosote	247-355	8.0
6	heavy oil	323-372 (90%)	9.5
residue	medium-soft pitch		56.6
liquor & losses			5.0

^a5-95% sidearm flask.

Source: McNeil, 1969.

TABLE 3. PREDOMINANT STRUCTURES IN COKE OVEN TAR

Boiling range (°C)	Average percent of tar	Major Components
0-150	0.8	Single 6-membered rings
		Benzene Toluene Xylenes
150-200	3	Fused 6,5-ring systems
		Indene Hydrindene Coumarone
200-250	12	Fused 6,6-ring systems
•		Naphthalene Methyl naphthalenes
250-300	8	Fused 6,6,5-ring systems
•		Acenaphthene Fluorene Diphenylene Oxide

Source: McNeil, 1966a.

Coal tar constituents by boiling point

≤100°C	Aniline Toluidine	4-Kethylindole
Pentane	2,3-Dimethylpyridine	5-Methylindole 7-Methylindole
rentane Hexane	2,3-Dimethylpyridine	3-Kethylquinoline
Heptane	2,5-Dimethylpyridine	4-Methylquinoline
neptane Cyclohexane	3,4-Dimethylpyridine	5-Methylquinoline
Butylene	Dimethylaniline	6-Methylquinoline
Amy lene	2,4,5-Trimethylpyridine	7-Methylquinoline
Hexene	2,4,6-Trimethylpyridine	l-Methylisoquinoline
Heptene	Benzonitrile	3-Methylisoquinoline
Cyclohexene		1,3-Dimethylisoquinoline
1.5-Butadiene	200 - 250°C	2,8-Dimethylquinoline
Crotonylene	Nanhaha Jago	5,8-Dimethylquinoline
Cyclopentadiene	Naphthalene Dihydronaphthalene	1-Naphthonitrile
Cyclohexadiene	a-Methylnaphthalene	· ·
Benzene	α-re try maph tha tene β-He thy lnaph tha lene	Above 300°C
Acetone	4.6-Dimethylindene	Nonadecane
Methylethyl ketone	5,7-Dimethylindene	Phenanthrene
Acetonitrile	m-Cresol	Anthracene
Carbon disulfide	p-Cresol	2-Methylfluorene
Methylmercaptan	1,2,3-Xylenol	3-Methylfluorene
Ethvimercaptan	1,2,4-Xylenol	Dihydroanthracene
Dimethyl sulfide	1,3,4-Xy lenol	4,5-Phenanthrylene methane
Diethyl sulfide	1,3,5-Xylenol	Chrysogen
Th i ophene	1,4,2-Xylenol	Methylanthracene
100 - 150°C	o-Ethylphenol	Fluoranthene
100 - 130 C	m-Ethylphenol	Pyrene
Octane	p-Ethylphenol	Tetrahydrofluoranthrene
Hethylcyclohexane	3-Methyl-5-ethylphenol	2,7-Dimethylanthracene
Dimethylcyclohexane	Isopseudocumenol	1,2-Benzfluorene
Nonaphthene	Dureno1	2.3-Benzfluorene
Toluene	2,2'-Dihydroxydiphenyl	Chrysene
o-Xylene	Acetophenone	1,2-Benzanthracene
m-Xylene	Benzoic acid	Naphthacene
p-Xylene	Dimethyl coumarone	Triphenylene
Ethylbenzene	1.2.3.4-Tetramethylpyridine	Perylene
Styrene	Quinoline	1,2-Benzpyrene
Pyrrole	Isoquinoline	Crackene
Pyridine	2-Methylquinoline	4,5-Benzpyrene
2-Methylpyridine	Thionaphthene	Picene
3-Methylpyridine	250 - 300°C	1,2-Benznaphthacene
4-Methylpyridine		Naphtho-2,3-1,2-anthracene
2,6-Dimethy1pyridine	Diphenyl	Acridine
Thiotoluene	3-Me thy I dipheny l	Diphenylene sulfide
Thioxene	2-Methyldiphenyl	Benzerythrene
150 - 200°C	4-Methyldiphenyl	1-Methylphenanthrene
	4,4'-dimethyldiphenyl	3-Methylphenanthrene
Cecane	3,4-Dimethyldiphenyl	9-Methylphenanthrene
Dicyclopentadiene	Acenaphthene	Di benzocoumarone .
Hydrindene	1,2-Dimethylnaphthalene	Carbazole
Isopropylbenzene	1,6-Dimethylnaphthalene	Benzacarbazole
o-Ethyltoluene	1,7-Dimethylnaphthalene	Truxene
::-Ethyltoluene	2.6-Dimethylnaphthalene	2-Methyldiphenylene oxide
p-Ethyltoluene	2,7-Dimethylnaphthalene	2-Hydroxyfluorene
n-Propylbenzene	2,3-Dimethylnaphthalene	1,9-Benzoanthrene 2-Naphthylamine
Mesitylene	2-Ethylnaphthalene	
Pseudocumene	1-Ethylnaphthalene	Hydroacridine
Hemellitene	1,2-Cyclopentanonaphthalene	2-Methylcarbazole
Cymene	Fluorene	3-Methylcarbazole
Durene	α-Naphthol	2-Phenylnaphthalene
Isodurene	β-Naphthol	p-Phenylphenol
Indene	Diphenylene oxide	2-Hydroxydiphenylene oxide
2,7-Dimethylindene	α-Naphthofurane	Phenanthridene
3.6-Dimethylindene	β-Naphthofurane	2-Phenanthrol
Phenol	l-Methyldiphenylene oxide	l-Naphthylamine
r-Cresol	Indo le .	2-Naphthonitrile
Coumarone	2-Methylindole	
Dimethylcoumarone	3-Methylindole	

Table 5

THE AMOUNTS OF THE MOST IMPORTANT COMPONENTS IN COKE

OVEN AND CONTINUOUS VERTICAL RETORT TARS

Components (% wt. on dry tar)	American Coke Oven Tars		
Benzene	0.12		
Toluene	0.25		
σ-Xylene	0.04		
m-Xylene	0.07		
p-Xylene ,	0.03		
Ethyl benzene	0.02		
Styrene	0.02		
Pheno1	0.61		
σ-Cresol	0.25		
m-Cresol	0.45		
p-Cresol .	0.27		
Xylenols	0.36		
High-boiling tar Acids	0.83		
Naphtha	0.97		
Naphthalene	8.80		
α-Methyl · naphthalene	0.65		
β-Methyl naphthalene	1.23		
Acenaphthene	1.06		
Fluorene	0.84		
Diphenylene oxide	¥-		
Anthracene	0.75		
Phenanthrene	2.66		
Carbazole	0.60		
Tar bases	2.08		
Medium-soft pitch	. 63.5		

Source: McNeil, 1966.

Table 6. Concentrations of PAH in coal tar and airborne coal tar emissions

Compound	Coal Tar Air Emissions		. Coal Tar	
• .	mg/g sample	ug/m ³ air sample	mg/g sample	
Fluorene	2.41	`85	27.39	
Dihydrophenanthrene and dihydroanthracene	0.68	24	1.99	
Methylfluorenes	0.74	26	3.12	
Phenanthrene and anthracene	142.90	5020	€81.07	
Methylphenanthrene and methylanthracene	2.18	77	2.53	
Ethylphenanthrene and ethylanthracene	3.88	136	4.36	
Octahydrofluoranthene	0.15	- 5	0.23 ·	
Octahydropyrene	0.15	5	0.21	
Dihydrofluoranthene and dihydropyrene	0.15	5	1.05	
Fluoranthene	144.85	5090	54.28	
Dihydrobenzofluorenes	8.85 .	311	5.26	
Pyrene	105.47	3705	46.50	
Benzofluorenes	24.53	862	20.13	
Methylfluoranthenes	3.91	137 -	2.25	
Methylpyrenes	3.88	136	2.05	
Trimethylfluoranthene and trimethylpyrene	1.29	45	0.66	
Benzo[ghi]fluoranthene	3.29	115	4.40	
Dihydrobenz[a]anthracene, dihydrochrysene and dihydrophenylene	2.65	93	3.87	
Benzo[a]anthracene, chrysene and triphenylene	45.74	1607	42.66	
Dihydromethylbenz[a]anthracene, chrysene and triphenylene	0.5	18	2.12	
Methylbenz[a]anthracene, methylchrysene and methyltriphenylene	4.26	150	11.04	
Dimethylbenz[a]anthracene, chrysene and triphenylene	0.41	15	1.87	
Benzo[j]fluoranthene	0.12	4	0.73	
Benzo[k]fluoranthene	9.96	350	. 32.54	

Table 6. (continued)

Compound	Coa Air Ei	Coal Tar		
Compound .	mg/g sample	μg/m ³ air sample	mg/g sample	
Methylbenzo[k]fluoranthene and methylbenzo[j]fluoranthene	2.47	: 88	7.43	
Benzo[a]pyrene and benzo[e]pyrene	13.83	486	33.13	
Methylbenzo[a]pyrene and methylbenzo[e]pyrene	2 . 15	76	8.76	
Dibenzanthracenes	0.14	14	2.13	
Benzo[b]chrysene and O-phenylenepyrene	3.88	137	22.79	
Benzo[ghi]perylene and anthanthrene	2.62	92	17.94	

Source: Lao et al., 1975.

Table 7

PAH in creosote and coal tar

DW4	Concentration in creosote (g/kg)		Concentration	Concentration in coal tar (g/kg)	
PN A	(1)	(2)	(g/liter)	(1)	(2)
Anthracene	12.1	12.0	6.2	2.88	4.35
Benz[a]anthracene	2.77	2.94	2.75	6.24	6.98
Benzo[b]chrysene	0.03	0.06		0.93	0.80
Benzo[j]fluoranthene	0.29	0.29		0.63	0.45
Benzo[k]fluoranthene	0.30	0.11		1.08	1.07
Benzo $[g,h,i]$ perylen $oldsymbol{e}$				1.23	1.89
Benzo[a]pyrene	0.14	0.22	0.12	2.08	1.76
Benzo[ø]pyrene	0.18	0.15		1.85	1.88
Carbazole	2.20	1.42	2.75	1.32	1.27
Chrysene	1.34	0.94	1.27	2.13	2.86
Dibenz[a , h]anthracene		-		0.30	0.23
Fluoranthene	24.8	22.2	7.8	17.7	17.8
Perylene	0.04	0.04	0.04	0.70	0.76
Phenanthrene	39.9	33.3	47.9	13.6	17.5
Pyrene	9.1	6.8	4.2	7.95	10.6

Source: Lijinsky et al. 1963, Table II, p. 954.

Table 8a Estimated Human Exposure to PAH from Various Sources ($\mu g/Day$)

BaP	Carcinogenic PAH	Total PAH	
0.0095-0.0435	0.038	0.207	
0.0011 المرام	0.0042 4.2pt	0.0270	- 27 ppt
0.16-1.6	b	1.6-16	
	0.0095-0.0435 0.0011	0.0095-0.0435 0.038 0.0011 - 191 0.0042 4-291	0.0095-0.0435

^aTotal of BaP, benzo(j)fluoranthene, and indeno(1,2,3-cd)pyrene.

Source: Santodonato et al., 1981.

^bNo data available.

Table 8b. Estimated inputs of benzo[a]pyrene and total PAH to the aquatic environment from various sources.

Sources .	Input in metric tons/year		
	BaP	Total PAH	
Biosynthesis	25	2,700	
Petroleum spillage	20-30	170,000	
Domestic and industrial wastes	29	4,400	
Surface runoff from land	118	2,940	
Fallout and Rain out from air	500	50,000	
Total input	697	230,040	

Source: Neff, 1979.

TABLE 9

PAH Concentration in Ground Water

		Concentration, µg/l		•
Source	BaP	Carcinogenic PAH	Total PAH	Reference
G. Finthen, Germany,		0.002		Borneff, 1964
Mainz, Germany		0.005		Borneff, 1964
Unspecified locations in Germany	0.0004	0.003	0.04	Borneff and Kunte, 1964
Average of 12 German ground waters*			0.06	Borneff and Kunte, 1969
Champaign, Ill.*	N.D.ª	0.003	0.007	Basu and Saxena, 1977
Elkhart, Ind.*	0.004	0.004	0.02	Basu and Saxena, 1977
Fairborn, 0.*	0.0003	0.0008	0.003	Basu and Saxena, 1977

^{*}These are results of 6 specified PAH

Source: EPA, 1980.

a_{N.D.:} not detected

	·	Concentration, µg/l		
Source	ВаР	Carcinogenic PAH	Total PAH	Reference
Rhine River at Mainz	0.08	0.49	1.12	Borneff and Kunte, 1964
River Main at Seligenstadt	0.0024	0.155	0.48	Borneff and Kunte, 1964
River Danube at Ulm	0.0006	0.067	0.24	Borneff and Kunte, 1964
River Gersprenz at Munster	0.0096	0.047	0.14	Borneff and Kunte, 1964
River Aach at Stockach	0.017	0.95	2.5	Borneff and Kunte, 1965
River Schussen	0.01	0.20	1.0	Borneff and Kunte, 1965
River Plyussa: at Shale-oil effluent discharge site 3,500 m	12			Dikun and Makhinenko, 1963
downstream	1			Dikun and
at Navy water intake	0.1			Makhinenko, 1963 Dikun and Makhinenko, 1963
A river: 15 m below coke by-product	8-12			Fedorenko, 1964
discharge site 500 m downstream	2-3		-	Fedorenko, 1964
Thames River at Kew Bridge	0.13	0.18	0.50	Harrison, et al. 1975
at Albert Bridge	0.16	0.27	0.69	Harrison, et al. 1975
at Tower Bridge	0.35	0.56	1.33	Harrison, et al. 1975

Source: .EPA, 1980.

TABLE 11 Concentrations of PAH in Raw and Treated Surface Water used as Drinking Water Sources

		<u> </u>	Concentration, pg/l		•		
Source	Treatment	BaP	Carcinogenic PAH	Total PAH	Reference		
River Rhine	Untreated	0.082	0.485	1.11	Borneff and Kunte, 1964		
River Rhine	Bank and acti- vated carbon filtered	0.0005	0.015	0.13	Borneff and Kunte, 1964		
Lake Constance	Untreated	0.0013	0.030	0.065	Borneff and Kunte, 1964		
Lake Constance ^a	Rapid sand filtration chlorination	0.0017	0.017	0.053	Borneff and Kunte, 1964		
English River	Untreated	0.06 ^b	0.37 ^G	0.73 ^b	Harrison, et al. 1976		
English River	Piltration and chlorination	0.009	0.051 ^G	0.24	Harrison, et al. 1976		
Monongahela River	Untreated	0.04	0.14	0.60	Basu and Saxena, 1978		
at Pittsburgh same as above	Treated ^d	0.0004	0.002	0.003	Basu and Saxena, 1978		
Ohio River at	Untreated	0.006	0.020	0.058	Basu and Saxena, 1978		
Huntington, W. Va. same as above	Treated	0.0005	0.002	0.007	Basu and Saxena, 1978		
Ohio River at	Untreated	0.21	0.57	1.59	Basu and Saxena, 1977		
Wheeling, W. Va. same as above	Treated ^d	0.002	0.011	0.14	Basu and Saxena, 1977		
Delawater River at	Untreated	0.04	. 0.16	0.35	Basu and Saxena, 1978		
Philadelphia same as above	Treated ^d	0.0003	0.002	0.015	Basu and Saxena, 1978		
Lake Winnebago at	Untreated	0.0006	0.002	0.007	Basu and Saxena, 1977		
Appleton, Wis. same as above	Treated ^đ	0.0004	0.002	0.006	Basu and Saxena, 1977		

Source: EPA, 1980.

These are average of five determinations with the exclusion of a sixth high value.

bThese values are estimates on the basis of average PAH adsorption in reservoir.

CThese values may be a little higher due to the inability of separation of all the carcinogenic from non-carcinogenic PAH.

The treatment included flocculation, activated carbon addition, filtration, pH control, chlorination and fluoridation.

PAH Levels in a Few Drinking Waters

	-	Concentration, ng/1		
Source	BaP	Carcinogenic PAH	Total PAH	Reference
Mixed tap water at Mainz, Germany			7.0	Borneff, 1964
Water at: a		,		
Syracuse, N.Y.	0.3	0.3	1.1	Basu and Saxena, 1978
Buffalo, N.Y.	0.2	0.2	0.9	Basu and Saxena, 1978
New York, N.Y.	0.5	3.9	6.4	Basu and Saxena, 1978
Lake George, N.Y.	0.3	1.5	4.2	Basu and Saxena, 1978
Endicott, N.Y.	0.2	1.1	8.3	Basu and Saxena, 1978
Hammondsport, N.Y.	0.3	1.5	3.5	Basu and Saxena, 1978
Pittsburgh, Pa.	0.4	1.9	2.8	Basu and Saxena, 1978
Philadelphia, Pa.	0.3	2.0	14.9	Basu and Saxena, 1978
Huntington, W. Va.	0.5	2.0	7.1	Basu and Saxena, 1978
Wheeling, W. Va.	2.1	11.3	138.5	Basu and Saxena, 1977
New Orleans, La.	1.6	1.6	2.2	Basu and Saxena, 1978
Appleton, Wis.	0.4	2.4	6.1	Basu and Saxena, 1977
Champaign, Ill,	N.D.b	1.2	2.8	Basu and Saxena, 1977
Fairborn, Ohio	0.1	0.8	2.9	Basu and Saxena, 1977
Elkhart, Ind.	и.р. ^b	0.3	0.3	Basu and Saxena, 1977

Only the six WHO (1970) - recommended PAH were analyzed, with the exception that BbFL replaced BjFl. PAH were concentrated by passing 60 liters of drinking water through polyurethane foams. The eluate from the foams was subjected to cleanup and analyzed for PAH by TLC-spectrofluorometry.

Source: EPA, 1980.

b_{N.D.:} not detected.

Table 13 Concentrations (ng/1) of PAHs in Surface and Ground Waters in Germany⁸

Location	FI	Руг	B(a)A	B(J)F	B(b)F	Ch	B(a)P	B(gh1)P	8(k)F	IP
Groundwater 1 May, 1963	42.0	b	b	b	0.8	b	0-1	0.8	0.8	0.4
Groundwater II August, 1963	26.2	b	1.0	1.0	1.0	b	0.6	0.5	0.5	0.5
Groundwater III January, 1964	169.0	104.0	23.2	10.0	11.5	b	23.4	17.5	10.0	12.6
Danube R. at Ulm April, 1964	94.0	74.5	11.0	10.1	24.2	b	0.6	9.5	7.7	9.5
Bodensee May, 1964	21.4	b	5.0	13.0	7.7	b	1.3	3.2	2.7	2.6
Danube R. at Ulm May, 1964	61.0	0.3	14.0	23.4	23.9	b	b	9.5	14-1	16.4
Main R. at Seligenstradt July, 1963	128.3	109.8	14.4	35.7	32.1	38.2	2.4	21.2	10.6	32.0
Main R. at Seligenstradt April, 1964	192.0	92.8	16.2	75.5	67.0	b	6.5	25.9	21.6	23.7
Rhine R. at Mainz May, 1964	146.0	b	53,5	21.3	77.8	b	49.2	43.2	27.4	35.8
Rhine R. at Mainz March, 1964	258.0	2.0	185.0	150.0	156.0	b	114.0	134.0	117.0	123.0

Table 14 $\,$ PAHs in Raw and Finished Waters in the United States a (ng/l)

	Cinci		New Orleans LA	Mia Fl		Seat VA		Portland OR	Columbus OH	Cape Gi M	rardeau O	Whee	ling	_	n Parish A	Tucso: AZ
Compound	Raw	Fin	Distributed	Raw	Fin	Raw	Fin	Raw	Fin	Raw	Fin	Raw	Fin	Raw	Fin	Raw
Ph	14	10	14	6	14	10	12	8.	3	14	5	9	4	20	14	10
FI .	<7	<3	<5	<4	<4	<4	<8	4	1	11	1	15	4	25	7	<2
Pyr	<14	<4	<3	<4	<6	<4	<6	6	<1	- 9	<1	15	2	18	3	<2
1-MP	<6	<4	b	<2	<1	<1	<1	<1	<1	<1	<1	5	<1	5	<2	<2
An	b	b	ь	Ь	ь	b	ь	b	b	1	<1	b	<1	2	<2	<2
Ch	4	<1	<1	<1	2	(1	2	<1	< 1	5	(1	8	<1	8	<2	<2
B(a)A	4	<1	< 1	<1	<1	<1	<1	< 1	<1	4	<1	9	- <1	9	<2	<2
Per	b	<1	<1	<1	<1	(1	<1	<1	<1	b	<1	<4	<1	7	b	b
B(e)P	b	<1	a	< 1	<1	<1	<1	<1	<1	b	<1	b	<1	<14	<2	<2
B(a)P	9	<1	<1	<1	<1	<1	<1	<1	<1	4	<1	13	<1	12	<2	<2
B(ghi)P	<1	<1	<1	<1	< 1	<1	<1	< 1	<1	4	(1	9	<1	7	b	b
B(b)F	5	<1	<1	<1	< 1	<1	<1	< 1	<1	4	<1	16	<1	9	<2	<2
B(k)F	3	<1	<1	<1	<1	a	<1	< 1	<1	2	<1	7	<1	3	<2	<2
DiB(ah)A	<1	<1	< 1	<1	<1	<1	<1	< 1	<1	(1	a	1	<1	<2	<2	<2
IP	<4	<1	< 1	<1	<1	<1	<1	<1	<1	<3	<1	<12	<1	∢ 9	<2	<2
Total	39	10	14	6	16	10	14	18	4	58	6	107	10	125	24	10

a Sorrell, et.al., 1979.
b Not analyzed.

Table 15. Relative half-lives in hours for degradation of PAH by some environmental oxidizers

Compound	RO ₂	Singlet Oxygen	0 ₃ (water) ^a	0 ₃ (air) ^b	C1 ₂	НО
Anthracene	38,000	5			•	
Dimethylanthracene		0.05		·	•	
Diphenylanthracene		0.6			t,<0.5 hr	t,∿10 h ¹⁵ for
Phenanthrene	2 x 10 ⁸				² for all PAH	all PAH
Pyrene	2.4×10^5		0.68	560		
Perylene	38,000					
Tetracene	96	<5				
Benzo(a)pyrene	2.4×10^{5}	5	1.05	870		
Benz(a)anthracene		10	0.45	370		
Dimethylbenz(a)anthracene		<5				
Dibenzanthracene			0.42	340		
Dimethyldibenzanthracene		<5	0.17	<140		
Rubrene		0.02				

a,
$$(0_3) = 10^{-4} \text{ M}$$

b, $(0_3) = 2 \times 10^{-9} \text{ M}$

Source: Radding, et al, 1976.

Table 16 . Rate constants and half-lives for reaction of PAH with ozone in water at $25\,^{\circ}\text{C}$

Compound	Rate Constant k ₂ , 1 mole ⁻¹ sec-1	t ₃₂ , minutes ^a	
Pyrene	.170	41	
Benzo[a]pyrene	110	63	
Benz[a]anthracene	260	27	
Dimethylbenzanthracene .	>680	<10	
Dibenzanthracene	280	25	

Calculated from $t_{1/2} = 0.69/k_{2}[0_{3}]$, where $[0_{3}] = 10^{-4} M$.

Source: Radding, et al., 1976.

Table 17. Photodegradation of PAH under natural light in mixed acetonewater or carbon tetrachloride-water solutions. Initial quantities of starting materials are normalized to 100 mg, except in the mixed 1-methylnaphthalene-heteroaromatic tests where equimolar quantities of starting materials were used

Compounds	Degree of Photodegredation (mg compound remaining)				
	5 hours	10 hours			
Anthracene	52.9	32.6			
Phenanthrene	57.0	35.9			
Benz[a]anthracene	45.5	0.0			
Chrysene	96.0	94.0			
Fluorene	94.3	91.9			
Pyrene	94.6	89.1			
Benzo[a]pyrene	93.6	90.5			
Benzothiophene +	181.0	36.3			
1-methylnaphthalene	0.0 ^a	0.0 ^a			
Dibenzothiophene +	20.6	16.2			
l-methylnaphthalene	40.1 ^a	0.0 ^a			

a. Quantity of naphthalene after photoirradiation. Source: Nagata and Kondo, 1977.

Table 18. Microbial degredation of ¹⁴C-PAH in water from a controlled ecosystem enclosure to which No. 2 fuel oil had been added

PAH (initial concentration)	Collection depth (m)	Incubation time (h)	Time after oil addition (days)	Degradation rate (µg/l/day_S.D.)	Turnover time (days)
enzo[a]pyrene (16 µg/l)	5-10	48	0	0	-
enzo[a]pyrene (16 µg/1)	5-10	24	3 .	1±0.7	1400
luorene (30 µg/1)	5-10	48	0	0	-
luorene (30 µg/l)	5-10	48	3	0	-
.ethylnaphthalene (50 μg/1)	0-5	24	0	10±6	500
lethylnaphthalene (50 µg/1)	0-5	24	3	. 26±4	200 .
aphthalene (50 µg/2)	0-5	24	0	10±3	500 .
laphthalene (50 µg/l)	0-5	16	3	250±7	22
aphthalene (50 µg/l)	` 0-5	10	4	100±5 :	57
Naphthalene (50 μg/l)	5-10*	10	4	500±11	10

Source: Lee and Takahashi, 1977.

Table 19. Concentration of PAH in water at various stages in a water purification plant

,		PAH Concenti	ration (µg/lite	r)
Compound	River Intake	After Reservoir	After Filtration	After Chlorination
Fluoranthene	0.150	0.140	0.081	0.045
Pyrene .	0.100	0.075	0.045	0.018
Benz[a]anthracene +				٠.
Chrysene	0.090	0.072	0.033	0.012
Benzo[b+j+k]fluoranthene	0.147	0.132	0.039	0.021
Benzo[a+e]pyrene	a	0.051	0.030	0.009
Perylene	a	0.039	0.024	0.006
Indeno[1,2,3-cd]pyrene	0.069	0.066	0.027	0.009
Benzo[ghi]perylene	0.072	0.063	0.033	0.009

a. Measurement impossible due to background.

Source: Harrison, et al., 1976.

Table 20 PAHs in W23

Analysis by MRI

	•	•
Compound	Water Samples (ng/l)	Tar Samples (mg/g)
naphthalene	130,000	8.8
acenaphthylene -	29,000	1.7
acenaphthene	17,000	5.8
fluorene	29,000	7.9
phenanthrene	49,000	27.0
fluoranthene	30,000	21.0
pyrene	14,000	14.0
chrysene	5,000	6.8
benzo(a)pyrene	9,000	5.9
benzo(b)fluoranthene	5,000	17.0
carbazole	11,000	0.54
benzo(ghi)perylene		3.9
acridine		0.72
1		

10/81

Table 21

A. PAHS IN WATER SAMPLES FROM W13

Analysis by	WRD
Compound	WRD of USGS (ng/l)
naphthalene	220,000
acenaphthylene	<5,250
acenaphthene	<5,250
phenanthrene	6,300
anthracene	6,600
fluoranthene	420,000
pyrene	500,000
chrysene	360
benz(a)anthracene	300,000
benzo(a)pyrene	160,000
dibenz(a,h)anthracene	160,000
indeno(1,2,3-C,d)pyrene	27,000
benzo(ghi)perylene	92,000

10/80

B. Amines & Heterocyclics in Water Samples from W13

2-methylpyridine	41
benzeneamine	705
4-methylbenzeneamine	647
3-methylbenzeneamine	297
2-methylquinolene	21
benzo(h)quinolene	7
acridine	106
benzo(f)quinolene	2
4-azafluorene	13

Table 22
PAHs IN WATER SAMPLES FROM SLP15

Compound Analy	rsis by Iowa (ng/l)	Analysis by WRD of USGS (ng/1)
naphthalene	1130	70
acenaphthylene	1475	<20
acenaphthene	780	35
fluorene	249	73
phenanthrene	113	110
anthracene	362	70
fluoranthene	532	70
pyrene	360	<6
benz(a)anthracene	9	<3
chrysene	2	<1
benzo(k)fluoranthene	1	•
benzo(a)pyrene	2	<5
dibenz(a,h)anthracene	3	<12
benzo(ghi)perylene	1	<7
	8/81	10/80

Table 23
PAHs IN WATER SAMPLES FROM SLP4

Compound	Analysis by Iowa (ng/l)	Analysis by WRD of USGS (ng/l)	Analysis by CH2M Hill (ng/l)
naphthalene	<10	<18	18
acenaphthylene	<60	<20	11
acenaphthene	107	<36	272
fluorene	27	<2	47
phenanthrene	<10	<1	84
anthracene	<31	<1	4
fluoranthene	6	<7	4
pyrene	10	<6	4
benz(a)anthracene	<1	<3	<1
į. chrysene	<2	<1	3
benzo(k)fluoranthene	<1		<1
benzo(a)pyrene	<1	<5	<1
n dibenzo(a,h)anthracene	<1	<12	<1
benzo(ghi)perylene	<1	<7	<1
indeno(1,2,3-c,d)pyrene		<2	<1
	8/81	10/80	5/82

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